Supporting Information

Hydronium Ion Acidity Above and Below the Interface of Aqueous Microdroplets

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Liquid microjets of pure acidified water or acidified water saturated with liquid isoprene at 298 K were injected into the reaction chamber of the electrospray ionization mass spectrometer (Agilent 6130 Quadrupole LC/MS Electrospray System at NIES) flushed with N_2(g) at 1 atm, 298 K. Gas-phase isoprene was injected in the chamber via a side port as a plume that crossed the liquid microjets orthogonally. Gas-phase isoprene was carried by flowing carrier gas over a liquid isoprene reservoir kept at 298 ± 3 K. The carrier gas was ultrapure (>99.999%) nitrogen gas flowing at 5–500 mL min^{-1} rates regulated by calibrated digital mass flow controllers (Horiba, STEC, SEC-400 MARK 3). We assumed that the carrier gas was saturated with isoprene vapor in equilibrium with the liquid. The conditions used in the present experiments were as follows: drying nitrogen gas flow rate: 12 L min−1; drying nitrogen gas temperature: 340 °C; inlet voltage: −3.5 kV relative to ground; and fragmentor voltage: 60 V. Isoprene (Tokyo Chemical Industries, ≥99%) was used as receive. Solution pH was adjusted by adding HCl and measured with a calibrated pH meter.